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Short communication

In-situ growth of needlelike $LaAl_{11}O_{18}$ for reinforcement of alumina composites

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Abstract

In situ growth of needlelike $LaAl_{11}O_{18}$ grains reinforcing Al_2O_3 composites can be fabricated by a coprecipitation method using $La(NO_3)_3{\cdot}6H_2O$ and $Al(NO_3)_3{\cdot}9H_2O$ as starting materials. The new two-step process involved firstly preparing needlelike $LaAl_{11}O_{18}$ grains distributed homogeneously in Al_2O_3 powder and then pressureless sintering the composite powders. The Al_2O_3/25 vol.%-$LaAl_{11}O_{18}$ samples pressureless sintered at 1550°C for 4 h achieve relative density up to 96.5% and exhibit a bending strength of 420 ± 30 MPa and a fracture toughness of 4.3 ± 0.4 MPa $m^{1/2}$.

Keywords: A. Grain growth; B. Platelets; D. Al_2O_3; In situ composite

1. Introduction

Because of its outstanding combination of high melting point, high electrical resistivity and chemical durability as well as its low cost availability, Al_2O_3 is among the most promising materials for use in structural, electrical, optical and biomedical applications. However, brittle failure under mechanical or thermal stress is a major limitation of Al_2O_3 in many applications [1]. Therefore, a major research objective has been to improve its toughness. One possible approach is to prepare composites by adding high aspect ratio reinforcements such as whiskers, fibers and platelets [2–4]. Conventional processes of fabricating whiskers, fibers and platelets reinforced Al_2O_3 matrix composites involve physically mixing Al_2O_3 materials with the reinforcements from a separate source. Generally, the second phases are mixed into Al_2O_3 powder by simply employing ball-milling and/or ultrasonic treatment. SEM observation showed that these processes can not achieve a homogeneous distribution of the second phases in the Al_2O_3 matrix, which degrade the mechanical properties of the Al_2O_3 matrix composites [5]. Moreover, the high cost of the reinforcements poses a significant barrier to commercialization and also the processing and handling of reinforcements are associated with severe health hazards [6]. A feasible, less-expensive and safer alternative is in situ formation and growth of highly anisotropic second-phase grains in Al_2O_3 composites [7].

In this paper, a new method for preparing in situ formation needlelike $LaAl_{11}O_{18}$ grains, reinforcing Al_2O_3 ceramic composites is proposed. First, in situ growth of needlelike $LaAl_{11}O_{18}$ in Al_2O_3 powder was synthesized by a coprecipitated method using $La(NO_3)_3{\cdot}6H_2O$ and $Al(NO_3)_3{\cdot}9H_2O$ as the starting materials and then the composites were fabricated by pressureless sintering the coprecipitated $Al_2O_3/LaAl_{11}O_{18}$ powders.

2. Experimental procedure

The $Al(NO_3)_3{\cdot}9H_2O$ and $La(NO_3)_3{\cdot}6H_2O$ were firstly dissolved in distilled water with a certain concentration, respectively and then the two solutions were mixed to yield a precipitate of net comosition of 25 vol.% $LaAl_{11}O_{18}$ and 75 vol.% Al_2O_3 by adjusting the pH value to 9–10 with $NH_3{\cdot}H_2O$ solution. After aging, the precipitate was filtered and washed twice with distilled water and ethanol, respectively. The gel was dried at 70–

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80°C for 24 h, after that the dried gel was milled with ethanol by adding AlF_3 in high purity alumina mediums and then again dried at 70–80°C for 12 h. The composite powders can be obtained by calcining the dried gel at 1500°C for 1 h, after that, they were dried and uniaxially pressed at 500 MPa and sintered at 1550°C for 4 h pressureless.

The phase composition of the powders was identified by X-ray diffraction (D/max-radiffractometer, Japan) with Ni filtered CuK_α radiation. The particle size and shape were characterized by SEM (EPMA-8705QHz, Japan) with energy dispersive X-ray analysis. The densities of the sintered samples were determined by the Archimedes method. A Vickers indenter (AKASHI) with 98 N load was employed to measure the fracture toughness. The room temperature bending strengths were measured in three-point bending with a span of 20 mm and a cross speed at 0.5 mm/min using an Instron-1195 Universal Test Machine. The dimension of the test piece was 2.5×5×25 mm.

3. Results and discussion

The dried gel calcined at 1500°C for 1 h was identified by XRD and is shown in Fig. 1. The XRD patterns for the powder showed well-defined peaks of $LaAl_{11}O_{18}$ and α-Al_2O_3. In this experiment, the AlF_3 additive was used to reduce the transformation temperature of the transition alumina to alpha alumina and enhanced the reactive activity of α-Al_2O_3 with La_2O_3 and $LaAlO_3$ [8]. So $LaAl_{11}O_{18}$ can be formed at a lower temperature which is slow or difficult to form by the conventional solid-state reaction technique alone [9]. The needlelike grains distributed homogeneously in the equiaxed grains can be seen from the SEM micrograph, as shown in Fig. 2(a). The combination of XRD and EPMA analysis indicated that the needlelike grains with aspect ratio of 3–5 are $LaAl_{11}O_{18}$ and the equiaxed grains are Al_2O_3 because of the needlelike grains (point A) containing elements La and Al and the equiaxed grains (point B) containing only element Al. The AlK_α and LaK_α peaks were clearly shown in Fig. 2(b). This processing takes a two-step process, because larger space in the loose composite powder, compared with the space in compacted green, is beneficial for rodlike $LaAl_{11}O_{18}$ grain growth.

The relative densities of the composites sintered pressureless at 1550°C for 4 h can reach 96.5% and the micrograph of Fig. 3 shows that some pores are present. The theoretical density of the Al_2O_3/25 vol.%$LaAl_{11}O_{18}$ composite estimated by the lever rule from the true density of each compound (Al_2O_3: 3.98 g/cm^3, $LaAl_{11}O_{18}$: 4.17 g/cm^3) is 4.03 g/cm^3 [10]. It is known that elongated second phases in matrix powders make it difficult to achieve full density [11]. However, in this experiment, the enhanced reactive activity of the powders obtained by the coprecipitated method and high shape-forming pressure are beneficial to improve the densities of the composites.

The bending strength and fracture toughness of the composites are shown in Table 1. The in situ development of needlelike $LaAl_{11}O_{18}$ grains in the Al_2O_3 matrix can improve the bending strength and fracture toughness. It is due to the formation of $LaAl_{11}O_{18}$ in the Al_2O_3 matrix which can inhibit the grain growth of Al_2O_3, leading to a more uniform and refined matrix microstructure, which improved the bending strength, and the

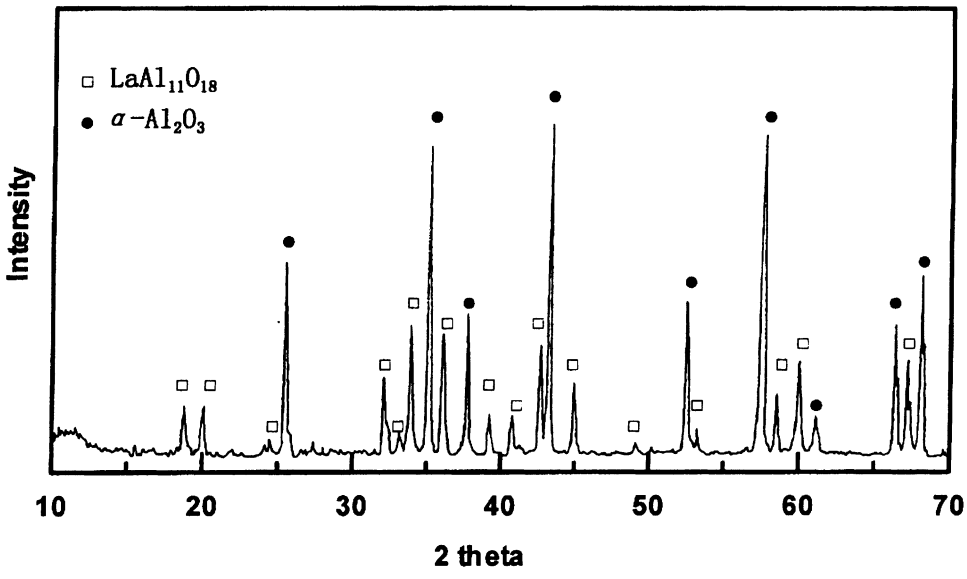

Fig. 1. XRD patterns of the powder calcined at 1500°C for 1 h.

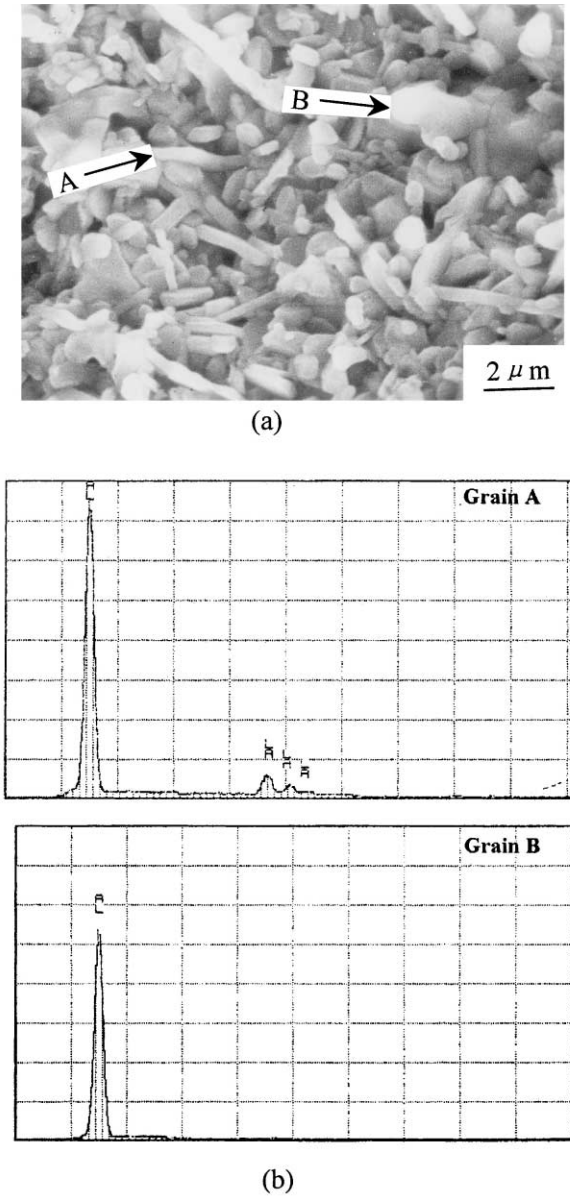

Fig. 2. (a) SEM micrograph of the composite powder; (b) EDS spectrum of points A and B.

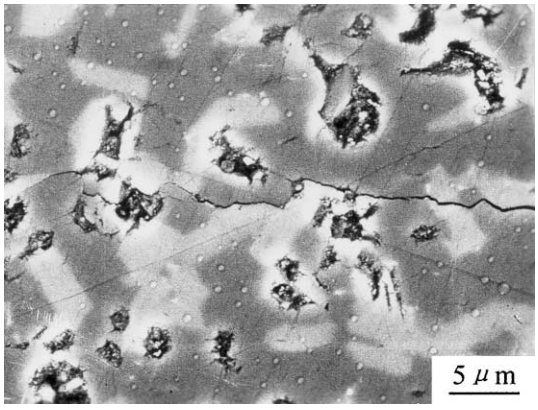

Fig. 3. SEM image of the crack propagation of the composites.

Table 1
Relative density, bending strength and fracture toughness of composites

Composition	Relative density (T.D. 4.03 g/cm^3)	Bending strength (MPa)	Fracture toughness (MPa m$^{1/2}$)
Al_2O_3/25 vol.% $LaAl_{11}O_{18}$	96.5	420±30	4.3±0.4

needlelike grains can also deflect the crack propagation, resulting in the increase of the fracture toughness. The SEM image of the crack traces introduced by the Vickers indentation on the surface of the composites is shown in Fig. 3, the distribution of the needlelike $LaAl_{11}O_{18}$ grains could be easily distinguished because they contain lanthanum, resulting in a bright image.

4. Conclusions

1. In situ growth of needlelike $LaAl_{11}O_{18}$ grains distributed homogeneously in Al_2O_3 powder can be synthesized by a coprecipitated method with $La(NO_3)_3{\cdot}6H_2O$ and $Al(NO_3)_3{\cdot}9H_2O$ as the starting materials.
2. $Al_2O_3/LaAl_{11}O_{18}$ composites can be sintered to 96.5% of theoretical density due to the enhanced activity of the coprecipitated powders and high shape-forming pressure.
3. Al_2O_3/25 vol.%$LaAl_{11}O_{18}$ composite sintered at 1550°C for 4 h exhibited a fracture toughness of about 4.3±0.4 MPa m$^{1/2}$ and a bending strength of around 420±30 MPa.
4. The SEM micrograph showed that the crack-deflection process operated as the main toughening mechanism in this composite.

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